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Effect of die surface on the onset of stick-slip flow in polymer processing

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Abstract. In this paper, we investigate the relationships between stick–slip defect and molecular slip at the wall for linear polymers. By considering Brochard–de Gennes slip model and molecular chain dynamics at the wall, we propose an expression for the critical shear stress at the onset of stick–slip as a function of the wettability of the polymer on the die surface or, more precisely, as a function of the apparent contact angle between polymer and die surface.

Introduction

Extrusion is one of the main processes in the thermoplastic industry. In this process, the polymer melt is pushed through a die to produce profiles with specific shapes (plaques, films, tubes...). At high extrusion rates, volume irregularities or surface defects appear on the surface of the extrudate. These instabilities (sharkskin, stick-slip...) are of the major concern in the polymer processing industry, limiting maximum production rates [1].

This work is focused on the instability observed for linear polyethylenes at high shear rates, called stick-slip or spurt. At constant flow rate in a capillary rheometer, this defect is characterized by pressure oscillations in the reservoir, which result in periodic flow at die exit. At constant pressure, the flow rate suddenly jumps to high values (spurt effect). To predict the onset of this phenomenon, we have considered the slip of the polymer melt at the die wall. Using the Brochard-de Gennes model [2], we have determined the critical value of stress for the onset of this defect, depending on rheological properties and molecular weight distribution [3]:

$$\sigma_c = \frac{9C_{ad}}{4\pi} \frac{G_N^0 N_e}{\sqrt{N_0}} \quad (1)$$

where G_N^0 is the plateau modulus, N_e the number of monomers between entanglements and N_0 the number of monomers per chain. The parameter C_{ad} takes into account the contacts between the adsorbed molecules and the die surface. It expresses the ratio of surface where molecules are adsorbed to the whole surface. In this work, we will express this coefficient as a function of the wettability of the polymer on the die surface or, more precisely, as a function of the apparent contact angle between polymer and die surface.

Contact angle

The basic parameter for a liquid wetting a solid is the contact angle as defined in Fig. 1 for a sessile drop. It is formed by a tangent to the liquid at the air/liquid/solid line of contact and a line through the base of the liquid drop where it contacts the solid. Almost universally, the angle reported is that measured through the drop as indicated in Fig. 1.

The lines of force acting along the line of intersection are the surface tensions of the liquid in equilibrium with the surrounding vapor γ_{LV} , the solid/vapor surface tension γ_{SV} and the solid/liquid surface tension γ_{SL} . Simple geometric considerations indicated in Fig. 1 lead to Young equation written in 1805 [4]:

$$\gamma_{SV} - \gamma_{LV} \cos \theta - \gamma_{SL} = 0 \quad (2)$$

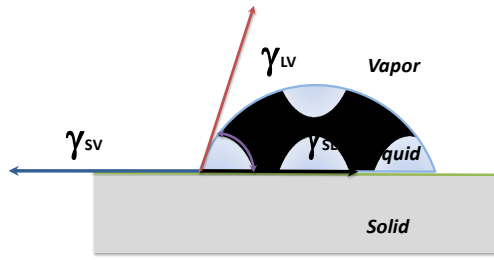


Fig. 1. Sessile drop profile showing contact angle θ and surface tensions acting at the three phase line of contact

The work of Zisman and co-workers [5-8] indicates that the contact angle of a liquid on a solid reflects in some way the chemical constitution of the solid surface. That could explain results on the effect of chemical nature of the die on the critical shear stress at the onset of the flow instabilities [9-11].

Work of adhesion

The notion of work of adhesion W_{adh} has been introduced by Dupré [12] in 1869 and constitutes the thermodynamic point of view on adhesion.

The Dupré equation gives the surface energy change that occurs when a unit area of liquid L is separated from a unit area of solid S:

$$W_{adh} = \gamma_s + \gamma_{LV} - \gamma_{SL} \quad (3)$$

where γ_s is the surface energy of the solid (in vacuum). Combining Young (Eq. (2)) and Dupré (Eq. (3)) equations gives:

$$W_{adh} = \gamma_{LV}(1 + \cos \theta) \quad (4)$$

The difference $\gamma_s - \gamma_{SL}$ is zero or essentially negligible. We have recently shown [13] that the coefficient C_{ad} could be expressed as:

$$C_{ad} = \frac{W_{adh}}{2\gamma_{LV}} \quad (5)$$

According to Eq. (4), C_{ad} can now be written as:

$$C_{ad} = \frac{1 + \cos \theta}{2} \quad (6)$$

Equation (6) shows how the contact fraction of polymer adsorbed on die surface is controlled by the wettability of the polymer melt.

Roughness effect

The surfaces considered in the Young equation (Eq. (1)) are supposed to be perfectly smooth and homogeneous and the state of the system is

an equilibrium state. In a real experimental situation, it is practically impossible to have such conditions; as a consequence, Young angle θ is an *ideal* quantity.

The requirement most often violated when measuring the equilibrium contact angle is surface uniformity, in terms of both roughness and chemical heterogeneity. There have been many attempts to relate the apparent contact angle θ_A to the equilibrium contact angle θ and the surface roughness. For Wenzel [14], the roughness increases both solid/liquid and solid/vapor interfacial tensions by a factor r , the surface roughness coefficient. The relation between θ_A and θ is:

$$r = \frac{\cos \theta_A}{\cos \theta} \quad (7)$$

Since r is always greater than 1, it follows that, when θ is less than 90° , θ_A increases with increasing θ . On the opposite, when θ is greater than 90° , θ_A decreases with increasing θ . Stated differently, if the equilibrium contact angle is less than 90° , the apparent contact angle decreases as the surface is roughened. On the other hand, if the equilibrium angle is greater than 90° , roughening of the surface causes the measured angle to increase. This is usually what is observed experimentally, although the situation can be more complex as discussed later.

In this condition, the coefficient C_{ad} is given by:

$$C_{ad} = \frac{1 + r \cos \theta}{2} \quad (8)$$

and the critical shear stress for the onset of stick-slip can be expressed as:

$$\sigma_c = \frac{9(1 + r \cos \theta)}{8\pi} \frac{G_N^0 N_e}{\sqrt{N_0}} \quad (9)$$

Confrontation with experiments

To compare the model with experimental data, we have selected works where were measured simultaneously both the contact angles and the critical shear stresses. We present on Figure 2 the comparison of the model predictions with the data of Anastasiadis and Hatzikiriakos [15] and Larrazabal et al. [10]. It shows a linear variation of the critical stress as function of the contact angle as predicted by Eq. (9) and confirms the results from the literature.

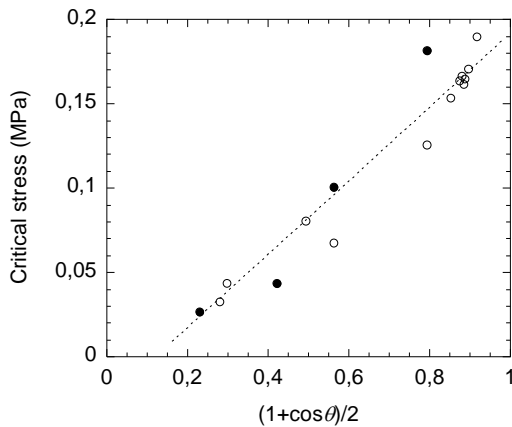


Fig. 2. Variation of the critical shear stress as function of the contact angle. Data from [10] (○) and [15] (●).

Conclusion

Based on work of adhesion and contact angle, we have proposed in this work a new expression for the critical shear stress for the onset of stick-slip. It takes into account the molecular structure of the polymer, the molecular weight distribution, the polymer/wall interactions and the effect of die wall roughness.

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